

Theory of Thermally-Stimulated Luminescence

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ABSTRACT

Thermoluminescence (TL) studies provide a new field of investigation of solid materials. The kinetic process is applicable to the theory of thermoluminescence (TL) and it is neither a first order kinetics nor a second order process. Instead an empirical general order equation has been proposed where the rate of escape of trapped electrons is proportional to the intensity of emitted light per unit time. Its applications have engulfed a whole spectrum of disciplines. The present paper reports the TL stimulated by different technique and discusses the correlation between mechanoluminescence (ML) and TL.

Keywords: Thermoluminescence (TL), Mechanoluminescence (ML), Trapped electrons.

INTRODUCTION

When ionizing radiations (X – rays, γ -rays, β particles and α particles) is incident upon an insulator or semiconductor and is heated, then the energy stored in the phosphor, as a result of irradiation process, liberates in the form of visible light in addition to the normal thermal radiation. The additional visible light emitted during first heating is called thermo luminescence. Reheating the phosphor, immediately after it gives rise to only normal thermal emission.

When an irradiated colored crystal is heated at a rapid rate, carriers (holes or electrons) are set free from the traps (defect sites) and emission takes place when they recombine with a charge of opposite sign. The defect sites which release the carriers are known as traps. In contrast, the centers from which thermal transitions are not likely but a probability of capture of a charge of opposite sign is appreciable, are called “recombination centers”. This emission is generally known as thermally stimulated luminescence or simply “thermo luminescence” abbreviated as TSL or TL.

The radiation dose absorbed by phosphor after the last heating of the sample is measured by the emitted TL. This dose can be estimated if the TL response to unit radiation dose (TL-sensitivity) is known. In most of the luminescence process, the exciting source is indicated by a prefix in the terminology, e.g., photoluminescence is excited by photons, but thermoluminescence is the misnomer because thermal energy is not the source for excitation. It is only a stimulant for material pre-excited by some other source of radiation to yield thermoluminescence. A theoretical basis to luminescence was given by¹⁻⁴.

Robert Boyle observation of a strange “glimmering light” on warming a diamond in the dark lead to discovery of TL is 1663 and he reported to the Royal Society in London. Most of the early work of this subject was restricted to natural minerals and glasses and it was only about three decades later when crystal growing techniques became common that the interest shifted to the TL phosphors grown in the laboratory. Although considerable works had been done before on TL in the context of solid state physics, it was in 1953 that the use of this phenomenon in measuring doses of ionizing radiations was first proposed by Farrington Daniels of the United States of America. Subsequently, John R. Cameron and his students in the Medical Physics Department of the University of Wisconsin made an intensive study on the development of lithium fluoride TLD phosphor⁵. In 1961, Harshaw Chemical Company (USA) began collaborating with Cameron’s group and in a couple of years, a new phosphor LiF: Mg, Ti known as TLD-200 was developed by them.

Later, Harshaw also made available lithium fluoride phosphors TLD-600 and TLD-700, which were composed of isotopically enriched lithium-6 and lithium-7, respectively. These three types of LiF materials are being used to-day.

The kinetic process is applicable to the theory of thermoluminescence (TL) and it is neither a first order kinetics nor a second order process. Instead an empirical general order equation has been proposed where the rate of escape of trapped electrons is proportional to the intensity of emitted light per unit time. TL is perhaps one of those rare physical phenomena which are more successfully applied than understood.

Thermoluminescence studies provide a new field of investigation of solid materials⁶⁻⁸. It can be a much more sensitive radiation detector than either a Geiger or Scintillation Counter. Its applications have engulfed a whole spectrum of disciplines such as Archaeology, Biology, Biochemistry, Forensic science, Geology, Radiation dosimetry, Radiation Physics, Solid State Physics, Space Science, Spectroscopic Analysis, TL Photography and so on. The most fundamental process in all these applications is the radiation dosimetry⁹⁻¹³.

We have studied the thermoluminescence stimulated by different technique, primarily to get help to understand the mechanism and characteristics of ML. The present paper reports the TL stimulated by different technique and discusses the correlation between ML and TL.

THEORY

The movement of electrons from the conduction band to the deeper levels which

provides the luminescence so $\frac{n_c}{\tau}$ is proportional to the light intensity I. The light intensity may given by

$$I = \eta n_o S \exp\left(-\frac{E}{kT}\right) \exp\left[-\int_{T_o}^T \frac{S}{\beta} \exp\left(-\frac{E}{kT}\right) dT\right] \quad (1)$$

where η is the probability that recombination will occur radiatively.

The first order characteristic that T_m independent on n_0 . The heating rate β must change T_m in such a way that the equally still holds. And β is given by

$$\beta = \left(\frac{Sk}{E}\right) T_m^2 \exp\left(-\frac{E}{kT_m}\right) \quad (2)$$

The maximum intensity I_m is given by

$$I_m = \eta S n_o \exp\left(\frac{-E}{kT_m}\right) \quad (3)$$

The nonlinear heating function $T(t)$ dependent intensity is given by

$$I(t) = \eta n_o S \exp\left(\frac{-E}{kT}\right) \exp\left\{-S \int_{T_o}^t \exp\left(\frac{-E}{kT(t)}\right) dt\right\} \quad (4)$$

If the heating function is monotonically increasing then

$$I(T) = \eta n_o S \exp\left(\frac{-E}{kT}\right) \exp\left[-S \int_{T_o}^T \left(\frac{dT}{dt}\right)^{-1} \exp\left(\frac{-E}{kT}\right) dT\right] \quad (5)$$

Similarly for the second order kinetics

$$I(T) = n_o^2 S' \exp\left(\frac{-E}{kT}\right) \quad (6)$$

We discuss electron traps and hole recombination centers.

The one governing the recombination is

$$I(t) = \frac{-dm}{dt} = A_m m n_c \quad (7)$$

where n_c is the concentration of free electrons.

The kinetics of the process in more general terms and to see how the simplified cases of first order, second order and more general case emerge in above equation.

These are related to the relation between the concentration of electrons in the conduction band and in traps and to the rate of change of these concentrations namely

$$\left|\frac{dn_c}{dt}\right| \ll \left|\frac{dn}{dt}\right|, n_c \ll n \quad (8)$$

The TL peak of a first order or second order kinetics a number of attempts have been made to add a third parameter is that of general order kinetics. According to which one assume that the glow peak is governed by

$$I = -\eta \frac{dn}{dt} = \eta S' n^b \exp\left(\frac{-E}{kT}\right) \quad (9)$$

where b is the order of kinetics.

The solution of above equation a linear heating with heating rate β is

$$I(t) = \eta S n_o \exp(-E/kT) \left\{ \frac{(b-1)S}{\beta} \int_{T_o}^T \exp(-E/kT) dT + 1 \right\}^{-b/(b-1)} \quad (10)$$

where $S = S' n_o^{b-1}$

The equation for mixed order kinetics is

$$I(t) = -\frac{dn}{dt} = S'n^2 \exp\left(\frac{-E}{kT}\right) + S'c_1 n \exp\left(\frac{-E}{kT}\right) \quad (11)$$

It is to be noted that this mixture is in the terms of the differential equation rather than in the first and second order solution as suggest empirically by¹⁴ showed that with the neutrality assumption one gets

$$I(t) = -\eta \frac{dn}{dt} = \eta S \exp\left(\frac{-E}{kT}\right) F(n) \quad (12)$$

where

$$F(n) = \frac{SA_n n(n+c_1)}{\{A_m(n+c_1) + A_n(N-n)\}} \quad (13)$$

The basic principle of laser stimulated TL is that the ionizing radiation generates electron-hole pairs. A fraction of this pairs recombine spontaneously, the radiative recombination results in fluorescence. The rest of them are captured at the trapping sites for electrons and holes. Some of these traps are thermally unstable and leak continuously giving rise to phosphorescence, while others are thermally stable at the storage temperature and do not significantly detrapp until stimulated externally. The phenomenon of thermally stimulated luminescence occurs when the detrapping take place due to heating of material and optically stimulated luminescence arises when the detrapping is caused by optical means i.e. The hole detrapping is also possible.

The fast detrapping rate enhances the signal to noise ratio. The intensity at the maximum thermally stimulated luminescence is given by

$$I_m = -n_0 S \exp(-E/kT_m) \exp \int_{T_0}^{T_m} \frac{S}{\beta} \exp(-E/kT) dT \quad (14)$$

where T_m is the maximum temperature.

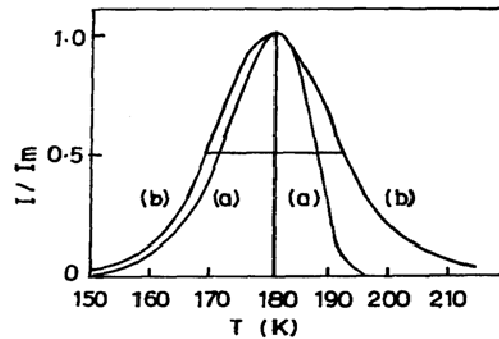


Fig. 1: TL peaks of first-order (curve a) and second-order (curve b) kinetics.

RESULTS AND DISCUSSION

Computed TL peaks of first-order (curve a) and second-order (curve b) kinetics. In the first-order peak, $E=0.4$ eV and $S=10^{10} \text{ sec}^{-1}$. In the second-order peak, $E = 0.4$ eV and $S=10^{11} \text{ m sec}^{-1}$. n_0 is chosen in curve b so that the two maxima coincide. Both curves are normalized. The important conclusion are drawn from the present investigation are as follows:

- (i) According to first order kinetics of TL following equations are derived

$$I = \eta n_0 S \exp\left(-\frac{E}{kT}\right) \exp\left[-\int_{T_0}^T \frac{S}{\beta} \exp\left(-\frac{E}{kT}\right) dT\right]$$

$$\frac{\beta E}{kT_m^2} = S \exp\left(-\frac{E}{kT_m}\right)$$

$$I_m = \eta S n_0 \exp\left(\frac{-E}{kT_m}\right)$$

$$I(t) = \eta n_0 S \exp\left(\frac{-E}{kT}\right) \exp\left\{-S \int_{t_0}^t \exp\left(\frac{-E}{kT(t)}\right) dt\right\}$$

$$I(T) = \eta n_o S \exp\left(\frac{-E}{kT}\right) \exp\left[-S \int_{T_o}^T \left(\frac{dT}{dt}\right)^{-1} \exp\left(\frac{-E}{kT}\right) dT\right]$$

(ii) According to second order kinetics of TL following equations are derived

$$I = S' n^2 \exp\left(\frac{-E}{kT}\right)$$

In the initial rise range i.e. T substantially below T_m then we get subsequently and the unity can be neglected so that one obtains.

$$I(T) \cong n_o^2 S' \exp\left(\frac{-E}{kT}\right) \left[\left(\frac{n_o S'}{\beta} \right) \int_{T_o}^T \left(\frac{-E}{kT} \right) dT \right]^{-2}$$

(iii) According to general order kinetics of a single TL peak following equations are derived

$$I(t) = \eta S n_o \exp\left(\frac{-E}{kT}\right) \exp\left\{ -\frac{S}{\beta} \int_{T_o}^T \exp\left(\frac{-E}{kT}\right) dT \right\}$$

$$I(T) = \eta n_o^2 S' \exp\left(\frac{-E}{kT}\right) \left[1 + \left(\frac{n_o S'}{\beta} \right) \int_{T_o}^T \exp\left(\frac{-E}{kT}\right) dT \right]^{-2}$$

$$I(t) = \eta S n_o \exp(-E/kT) \left\{ \frac{(b-1)S}{\beta} \int_{T_o}^T \exp(-E/kT) dT + 1 \right\}^{-b/(b-1)}$$

$$I(t) = -\eta \frac{dn}{dt} = \eta S \exp\left(\frac{-E}{kT}\right) F(n)$$

where

$$F(n) = \frac{S A_n n(n + c_1)}{\{A_m(n + c_1) + A_n(N - n)\}}$$

$$I(T) \approx n_o^2 S' \exp\left(\frac{-E}{kT}\right)$$

At higher temperature the latter term increases

(iv) Laser stimulated TL intensity are derived

$$I_m = -n_o S \exp(-E/kT_m) \exp \int_{T_o}^{T_m} S / \beta \exp(-E/kT) dT$$

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